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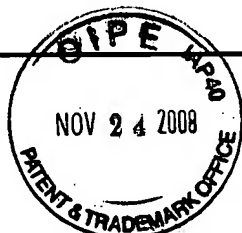
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**September 19, 2005**

Examiner

**EGWIM, Kelechi Chidi**

Group Art Unit

**1796**Invention: **AUXILIARY FOR FORMING FINE PATTERN AND PROCESS FOR PRODUCING THE SAME**I hereby certify that this **Machine Language English Translation from JPO of JP 9-235318 A - 25 Pages***(Identify type of correspondence)*

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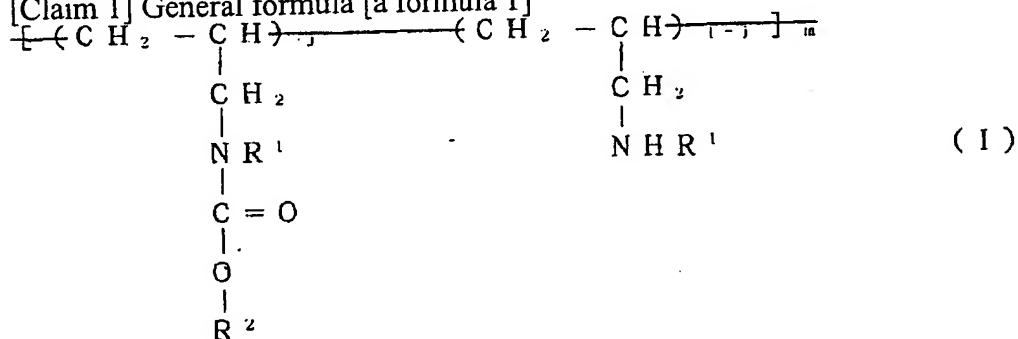
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## CLAIMS

[Claim(s)]

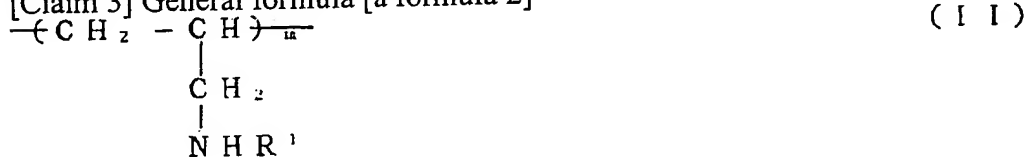
[Claim 1] General formula [a formula 1]



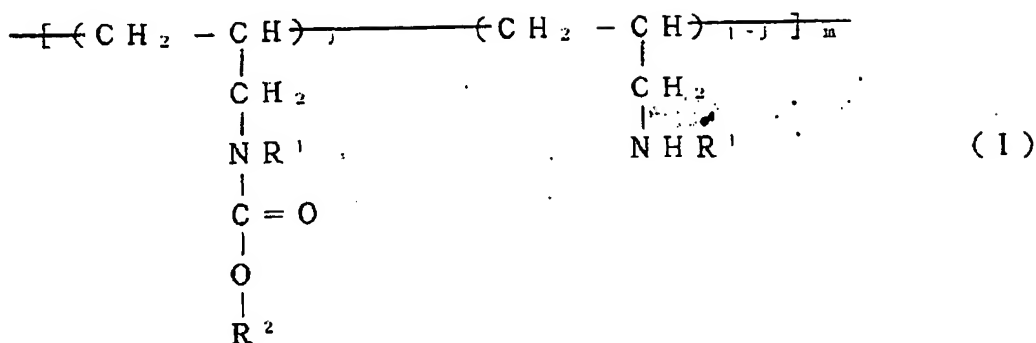
(m shows ten or more integers among a formula, and j shows the number with which are satisfied of  $0 < j \leq 1$ .) R1 shows n- of hydrogen and carbon numbers 1-8 and an iso-alkyl group, and the substituent chosen as a list from the cycloalkyl radical of carbon numbers 5-12. R2 the substituent chosen from the alkyl group and aryl group of carbon numbers 1-12 -- being shown -- salt of N-allyl compound urethane system polymer expressed or its N-allyl compound urethane system polymer.

[Claim 2] R1 It is hydrogen and is R2. Salt of N-allyl compound urethane system polymer according to claim 1 which is the substituent chosen from n- of carbon numbers 1-12, and an iso-alkyl group, or its N-allyl compound urethane system polymer.

[Claim 3] General formula [a formula 2]



In the partial salt of the poly allylamine system polymer expressed with (m shows ten or more integers among a formula, and R1 shows the substituent chosen as n- of hydrogen and carbon numbers 1-8, and an iso-alkyl group list from the cycloalkyl radical of carbon numbers 5-12), or its poly allylamine system polymer The general formula characterized by making the carbonic acid diester expressed with general formula R2 O-CO-OR2 (the substituent as which R2 is chosen from the alkyl group and aryl group of carbon numbers 1-12 among a formula is shown) react [a formula 3]



(m shows ten or more integers among a formula, and j shows the number with which are satisfied of  $0 < j \leq 1$ .) R1 shows the substituent chosen as a hydrogen list from the cycloalkyl radical of carbon numbers 5-12 by n- of carbon numbers 1-8, and the iso-alkyl group list. R2 the substituent chosen from the alkyl group and aryl group of carbon numbers 1-12 -- being shown -- the manufacture approach of the salt of N-allyl compound urethane system polymer expressed or its N-allyl compound urethane system polymer.

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[Translation done.]

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## DETAILED DESCRIPTION

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### [Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the new poly allylamine system derivative and its manufacture approach. Furthermore, it is related with new N-allyl compound urethane system polymer with which the amino group has a reactant side chain few in detail, and its easy manufacture approach.

[0002]

[Description of the Prior Art] The poly allylamine and a poly allylamine system polymer like Pori (N-alkyl allylamine) are cation system macromolecules which are the olefin system polymers of the straight chain containing the amino group, melt into a side chain in water well, and carry out an electric charge to plus underwater. Such a poly allylamine system polymer has peculiar reactive polymer structure and a peculiar property, therefore is used for the color binder for reactive dye, the color binder for direct dye, the food preserver, the anchor coat agent, etc. Moreover, using the poly allylamine system polymer in fields, such as silver halide photosensitive material, a sustained-release physic constituent, ion exchange resin, and functional film, is proposed. However, if it generally says as indicated by the "composition [ of a reactant polymer ], and application" 80-92 page (1989) of the CMC issue, the number of the reaction radicals of a reactive polymer does not have not much many a certain need, and even when inconvenient [, and ], there is. [ too ] Although, compounding the polymer which decreased the amino group by copolymerization on the other hand is also considered, since monoallyl amine system monomers, such as a monoallyl amine and monochrome (N-alkyl allylamine), are not copolymerized, as for the usual vinyl system monomer, it is difficult monomers to compound the poly allylamine system derivative of a low cation consistency by copolymerization. Then, how to make the amino group of the poly allylamine system polymer react with other compounds, and change into a deactivating group is also examined. The allylurea polymer (JP,63-43403,B) is known as a poly allylamine system derivative manufactured by such approach. However, the present condition is this poly allylamine system derivative's also having problems, like it being difficult to refine, and hardly used practical.

[0003] On the other hand, since the poly allylamine system polymer melts in water well, the application of the adhesion fields, such as a drainage system anchor coat agent (JP,4-292640,A) as an alternative of an organic solvent system anchor coat agent, is known. However, there was a practical problem that a water resisting property was missing in the laminate film produced using these anchor coat agents. Therefore, development of the new poly allylamine system derivative whose water resisting property crosslinking reaction occurs easily, and insolubilizes by heat-treatment, consequently improves was desired.

[0004]

[Problem(s) to be Solved by the Invention] It is offering the approach of manufacturing simply N-allyl compound urethane system polymer and it which as for the purpose of this invention it has a reactant side chain, and crosslinking reaction's occurs easily by heat-treatment further, and can insolubilize there being little amino group.

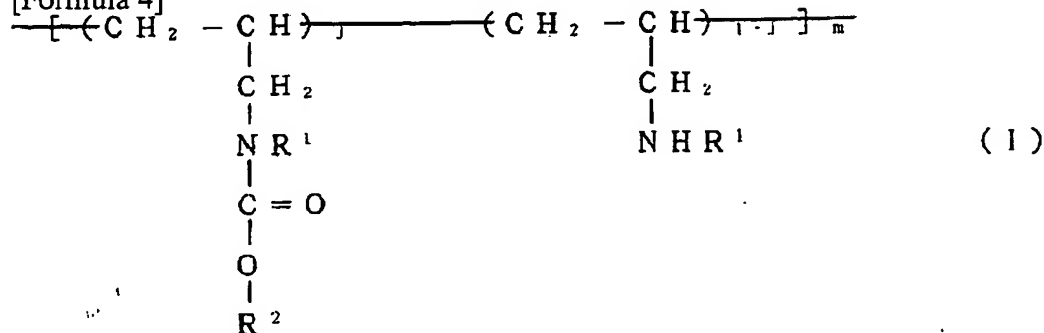
[0005]

[Means for Solving the Problem] By making the specific poly allylamine system polymer and specific carbonic acid diester react, there is little amino group, and it finds out manufacturing simply the new poly allylamine system polymer which has a block isocyanate radical, and this invention persons came to make this invention based on this knowledge, as a result of the amino group's repeating research wholeheartedly about the approach of manufacturing simply the new poly allylamine system derivative and new it which have a reactant side chain few. In addition, on these specifications,

urethane-ization means that the amino group (NH) changes to an alkyloxy carbonylamino radical or an aryloxy carbonylamino radical (N-CO-OR<sub>2</sub>).

[0006] This invention is a general formula [0007].

[Formula 4]



(m shows ten or more integers among a formula, and j shows the number with which are satisfied of  $0 < j \leq 1$ .) R<sub>1</sub> shows n- of hydrogen and carbon numbers 1-8 and an iso-alkyl group, and the substituent chosen as a list from the cycloalkyl radical of carbon numbers 5-12. R<sub>2</sub> the substituent chosen from the alkyl group and aryl group of carbon numbers 1-12 -- being shown -- it is the salt of N-allyl compound urethane system polymer expressed or its N-allyl compound urethane system polymer.

[0008] N-allyl compound urethane system polymer of this invention has a block isocyanate radical. Therefore, under heating conditions, since urethane becomes isocyanate, reactivity usually becomes high.

[0009] The salt of N-allyl compound urethane system polymer of this invention and its N-allyl compound urethane system polymer is the stability when making it the stability when making it a solution, especially an acidic solution, the solubility over moderate water, and the ease of receiving of a raw material to R<sub>1</sub>. Hydrogen and R<sub>2</sub> What is n- of carbon numbers 1-12 or an iso-alkyl group is desirable.

[0010] In this invention, m expresses the polymerization degree of N-allyl compound urethane system polymer of this invention. m is 10-5000 preferably ten or more.

[0011] In this invention, j expresses whenever [ urethane-ized ]. Although j is  $0 < j \leq 1$ ,  $0.05 \leq j \leq 1$  is usually desirable from whenever [ reduction / of the cation consistency of N-allyl compound urethane system polymer of this invention ].

[0012] It sets to this invention and is R<sub>1</sub>. The substituent chosen as n- of hydrogen and carbon numbers 1-8 and an iso-alkyl group list from the cycloalkyl radical of carbon numbers 5-12 is shown. It is R<sub>1</sub> practically from the ease of carrying out of the solubility over moderate water, and acquisition of a raw material. Hydrogen is desirable.

[0013] It sets to this invention and is R<sub>2</sub>. The substituent chosen from the alkyl group and aryl group of carbon numbers 1-12 is shown. In this case, methyl, ethyl, and propyl can be illustrated as an alkyl group of carbon numbers 1-12. R<sub>2</sub> As an aryl group of carbon numbers 1-12, a phenyl group, o-tolyl group, m-tolyl group, p-tolyl group, etc. can be illustrated. The stability when making it into the stability when making it a solution, especially an acidic solution in N-allyl compound urethane system polymer of this invention to R<sub>2</sub> It is desirable that it is n-alkyl group or the iso-alkyl group of carbon numbers 1-12.

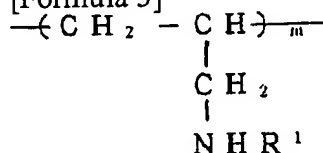
[0014] In the salt of N-allyl compound urethane system polymer of this invention, a salt is an addition salt to the side-chain amino group in the monomeric unit which is not urethane-ized. As such an addition salt, hydroxy acid salts, such as carboxylate, such as a hydrochloride, the hydrobromate, an iodine hydro acid salt, a sulfate, phosphate, phosphonate, and acetate, a methansulfonic acid salt, a p-toluenesulfonic-acid salt, citrate, and a tartrate, and a benzoate can be illustrated.

[0015] Generally, when N-allyl compound urethane system polymer of this invention is made into a salt, it may become unstable in the state of a solution. Therefore, in the state of a solution, the free thing of N-allyl compound urethane system polymer of this invention is desirable.

[0016]

[Embodiment of the Invention] N-allyl compound urethane system polymer of this invention can make the carbonic acid diester of the specific poly allylamine system polymer and specification able to react, and can be manufactured. That is, N-allyl compound urethane system polymer of this invention is a general formula [0017].

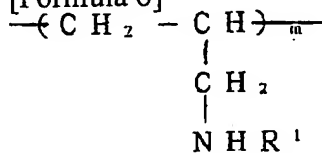
[Formula 5]



( I I )

the inside of a formula, and m -- ten or more integers -- being shown -- R1 -- n- of hydrogen and carbon numbers 1-8, and an iso-alkyl group -- the substituent chosen as a list from the cycloalkyl radical of carbon numbers 5-12 -- being shown -- to the poly allylamine system polymer expressed It can manufacture by making the carbonic acid diester expressed with general formula R2 O-CO-OR2 (the substituent as which R2 is chosen from the alkyl group and aryl group of carbon numbers 1-12 among a formula is shown) react. The poly allylamine system polymer of a raw material for manufacturing N-alkyl compound urethane system polymer of this invention is a general formula [0018].

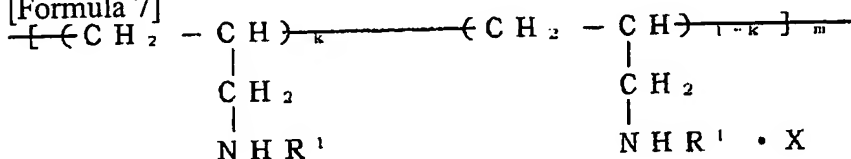
[Formula 6]



( I I )

It is expressed with (m shows ten or more integers among a formula, and R1 shows n- of hydrogen and carbon numbers 1-8 and an iso-alkyl group, and the substituent chosen as a list from the cycloalkyl radical of carbon numbers 5-12). [0019] Moreover, the partial salt of the poly allylamine system polymer can also be used as such a raw material. Such a partial salt is [0020].

[Formula 7]



( I I I )

It is expressed as a partial salt expressed with (m shows ten or more integers among a formula, k shows the number with which are satisfied of  $0 < k < 1$ , and R1 shows n- of hydrogen and carbon numbers 1-8 and an iso-alkyl group, and the substituent chosen as a list from the cycloalkyl radical of carbon numbers 5-12). In this case, although X is an acid for forming a partial salt, if it is the acid which can form the amino group and salt of a side chain of the poly allylamine system polymer, it will not be limited especially. As such an acid, hydroxy acid, such as carboxylic acids, such as a hydrochloric acid, a hydrobromic acid, iodine hydro acid, a sulfuric acid, a phosphoric acid, phosphonic acid, and an acetic acid, methansulfonic acid, p-toluenesulfonic acid, a citric acid, and a tartaric acid, and a benzoic acid can be illustrated.

[0021] The poly allylamine system polymer of a raw material is R1. It is the poly allylamine when it is hydrogen. The free type thing of the poly allylamine is desirable. Although what neutralized the salt of the known poly allylamine with alkali can be used as a free type commercial 15% poly allylamine water solution (the Nitto Boseki Co., Ltd. make --) of molecular weight about 10,000 concentration 10% poly allylamine water solution (the Nitto Boseki Co., Ltd. make --) of PAA-15 and molecular weight about 10,000 concentration PAA-10C, 20% poly allylamine water solution (the Nitto Boseki Co., Ltd. make, PAA-L) of molecular weight about 10,000 concentration, 20% poly allylamine water solution (the Nitto Boseki Co., Ltd. make, PAA-H) of molecular weight about 100,000 concentration, etc. may be used as it is.

[0022] It sets to the poly allylamine system polymer of a raw material, and is R1. When it is n- of carbon numbers 1-8 and an iso-alkyl group, and the substituent chosen as a list from the cycloalkyl radical of carbon numbers 5-12 By approach which is indicated by JP,63-43402,B, JP,6-2779,B, and JP,2-57084,B The salt of Pori (N-alkyl allylamine) can be manufactured and what removed the neutralization salt which subsequently carries out the byproduction of the water solution of the salt of the Pori (N-alkyl allylamine) after neutralizing with alkali, for example, a sodium hydroxide, for example, a sodium chloride, by dialysis can be used.

[0023] As a solvent for using the poly allylamine system polymer of a raw material as a solution, the mixed solvent of water, an organic solvent, or a water and an organic solvent can be used. As an organic solvent, the solubility of a raw material to a polar solvent is desirable, and alcohols, such as a methanol, ethanol, 1-propanol, 2-propanol, and 1-butanol, an acetonitrile, a formamide, N,N-dimethylformamide, dimethyl sulfoxide, a tetrahydrofuran, 1,4-dioxane, etc. can be illustrated. As for the amount of the solvent for using the poly allylamine system polymer of a raw material as a solution, it is desirable to use it 0.5 to 100 weight to poly allylamine system polymer 1 weight of a raw material.

[0024] The carbonic acid diester of a raw material for manufacturing N-allyl compound urethane system polymer of this invention is expressed with general formula R2 O-CO-OR2 (R2 shows among a formula the substituent chosen from the alkyl group and aryl group of carbon numbers 1-12). As carbonic acid diester, dimethyl carbonate, diethyl carbonate, diphenyl carbonate, etc. can be illustrated. The amount of the carbonic acid diester made to react can usually use required computational complexity for whenever [ urethane-ized / of N-allyl compound urethane system polymer ] at stoichiometric.

[0025] When making the poly allylamine system polymer and carbonic acid diester of a raw material react and manufacturing N-allyl compound urethane system polymer of this invention, it is good to drop carbonic acid diester at the solution of the poly allylamine system polymer of a raw material slowly first. At this time, carbonic acid diester can be dissolved in a solvent and it can also be dropped at the solution of the poly allylamine system polymer of a raw material. In this case, the solvent for dissolving carbonic acid diester is usually the same as the solvent for dissolving the poly allylamine system polymer of a raw material. As for the reaction of the poly allylamine system polymer and carbonic acid diester, it is desirable to carry out stirring. Reaction temperature is preferably good to maintain [ 0-100-degree C ] at 30-60 degrees C still more preferably. When reaction temperature is too high, the generated urethane may decompose. Reaction time is 12 - 25 hours preferably, and can usually obtain the solution of the allyl compound urethane polymer of this invention for 12 to 48 hours. In order to remove the alcohol and the reaction solvent which carried out the byproduction after reaction termination, N-allyl compound urethane system polymer of this invention can be obtained as a solid-state by carrying out the vacuum drying of the reaction solution. A vacuum drying is good for temperature to dry by 25-70 degrees C and the vacua desirable and suitable at 35-60 degrees C, when dimethyl carbonate, diethyl carbonate, or carbonic acid dipropyl is used as water and carbonic acid diester as a solvent. When temperature is too high, side reaction may start. Moreover, when manufacturing the salt of N-allyl compound urethane system polymer of this invention, this and carbonic acid diester can be manufactured by making it react like the case where the free poly allylamine system polymer is used, using the partial salt of the poly allylamine system polymer as a raw material. usually, the case where the partial salt and carbonic acid diester of the poly allylamine system polymer of a raw material are made to react -- the poly allylamine system polymerization -- NH which does not form the salt is preferentially urethane-ized by NH in the living body. It becomes possible to take out the salt of N-allyl compound urethane system polymer of this invention as a solid-state by adding the solution of the salt of N-allyl compound urethane system polymer obtained to solvents, such as an acetone, and reprecipitating it after reaction termination.

[0026] It depends for whenever [ urethane-ized / of N-allyl compound urethane system polymer of this invention ] (mol %) on the amount of the carbonic acid diester of the used raw material. When the carbonic acid diester of an equimolecular amount is used to the amino group of the poly allylamine system polymer of a raw material, the amino group is usually almost urethane-ized. Therefore, the cation consistency of N-allyl compound urethane system polymer of this invention can be adjusted by adjusting the amount of the carbonic acid diester used as a raw material. Moreover, the hydrophobicity of N-allyl compound urethane system polymer of this invention can be changed according to the class of carbonic acid diester to be used. Therefore, when using N-allyl compound urethane system polymer of this invention for various applications, what has a suitable cation consistency and has suitable hydrophobicity can be chosen.

[0027] Furthermore, N-allyl compound urethane system polymer of this invention can choose the dissociation temperature of a block isocyanate radical by choosing the urethane group to introduce from being also the poly allylamine which has a block isocyanate radical.

[0028] Below, an example shows N-allyl compound urethane system polymer and its manufacture approach of this invention. As for this invention, it is needless to say that it is not what is limited by these examples.

[0029] example 1 20-mol % -- the manufacture agitator of methoxycarbonyl-ized poly allylamine (they are  $j=0.2$ ,  $R1=H$ , and  $R2=CH3$  with N-allyl compound urethane system polymer I of this invention) -- To the 300ml separable flask equipped with the tap funnel, the thermometer, and the reflux condenser 10.1% poly allylamine water solution (the

Nitto Boseki Co., Ltd. make --) of molecular weight about 10,000 concentration PAA-10C200g (it is 0.35 mols at the monomeric unit of the poly allylamine) was put in, temperature was kept at 50 degrees C, 6.38g (product made from MITEKKUSU) (0.07 mols) of dimethyl carbonate was hung on the water solution for 15 minutes, and it was dropped at it. While after dropping termination kept temperature at 50 degrees C, the reaction was continued for 12 hours. The reaction solution of transparent light yellow was obtained. pH of this solution was set to 10.84. next, the desiccating agent after taking 1g of this reaction solution to a weighing bottle and carrying out a vacuum drying at a room temperature for 24 hours -- phosphoric oxide -- using -- 50 degrees C -- a 48-hour vacuum drying -- carrying out -- 20-mol % -- methoxycarbonyl-ized poly allylamine was obtained. The results of elemental analysis were C= 58.97%, H= 10.93%, and N= 19.76%. These values were in agreement with C= 59.44% of calculated value, H= 10.86%, and N= 20.39%.

[0030] example 2 60-mol % -- in the same reaction container as the manufacture example 1 of methoxycarbonyl-ized poly allylamine (they are  $j=0.6$ ,  $R1=H$ , and  $R2=CH_3$  with N-allyl compound urethane system polymer I of this invention) 10.1% poly allylamine water solution (the Nitto Boseki Co., Ltd. make --) of molecular weight about 10,000 concentration PAA-10C200g (it is 0.35 mols at the monomeric unit of the poly allylamine) was put in, temperature was kept at 50 degrees C, 19.14g (product made from MITEKKUSU) (0.21 mols) of dimethyl carbonate was hung on the water solution for 40 minutes, and it was dropped at it. While after dropping termination kept temperature at 50 degrees C, the reaction was continued for 12 hours. The reaction solution of transparent light yellow was obtained. pH of this solution was set to 9.78. 1g of next, this reaction solution -- a weighing bottle -- taking -- the same conditions as an example 1 -- drying -- 60-mol % -- methoxycarbonyl-ized poly allylamine was obtained. The results of elemental analysis were C= 54.32%, H= 9.17%, and N= 14.75%. These values were in agreement with C= 54.88% of calculated value, H= 8.99%, and N= 15.24%.

[0031] drawing 1 -- 60-mol % -- the infrared absorption spectrum of methoxycarbonyl-ized poly allylamine was shown. An absorption spectrum has absorption of 1580cm<sup>-1</sup> based on the first amino group (-NH<sub>2</sub>), and 1700cm<sup>-1</sup> based on C=O of a urethane group (N-CO-O-R).

[0032] example 3 100-mol % -- in the same reaction container as the manufacture example 1 of methoxycarbonyl-ized poly allylamine (they are  $j=1$ ,  $R1=H$ , and  $R2=CH_3$  with N-allyl compound urethane system polymer I of this invention) 10.1% poly allylamine water solution (the Nitto Boseki Co., Ltd. make --) of molecular weight about 10,000 concentration PAA-10C200g (it is 0.35 mols at the monomeric unit of the poly allylamine) was put in, temperature was kept at 50 degrees C, 33.50g (product made from MITEKKUSU) (0.37 mols) of dimethyl carbonate was hung on the water solution for 1 hour, and it was dropped at it. While after dropping termination kept temperature at 50 degrees C, the reaction was continued for 12 hours. The reaction solution of transparent light yellow was obtained. pH of this solution was set to 7.85. 1g of next, this reaction solution -- a weighing bottle -- taking -- the same conditions as an example 1 -- drying -- 100-mol % -- methoxycarbonyl-ized poly allylamine was obtained. The results of elemental analysis were C= 51.68%, H= 7.94%, and N= 11.83%. These values were in agreement with C= 52.16% of calculated value, H= 7.88%, and N= 12.17%.

[0033] example 4 20-mol % -- in the same reaction container as the manufacture example 1 of ethoxycarbonyl-ized poly allylamine (it is  $j=0.2$ ,  $R1=H$ , and  $R2=C$  two H5 with N-allyl compound urethane system polymer I of this invention) 10.1% poly allylamine water solution (the Nitto Boseki Co., Ltd. make --) of molecular weight about 10,000 concentration PAA-10C200g (it is 0.35 mols at the monomeric unit of the poly allylamine) was put in, temperature was kept at 50 degrees C, 8.37g (a best, Nakarai Tesuku make) (0.07 mols) of diethyl carbonate was hung on the water solution for 15 minutes, and it was dropped at it. While after dropping termination kept temperature at 50 degrees C, the reaction was continued for 24 hours. The reaction solution of transparent light yellow was obtained. pH of this solution was set to 10.88. 1g of next, this reaction solution -- a weighing bottle -- taking -- the same conditions as an example 1 -- drying -- 20-mol % -- ethoxycarbonyl-ized poly allylamine was obtained. The results of elemental analysis were C= 59.94%, H= 10.79%, and N= 19.32%. These values were in agreement with C= 60.47% of calculated value, H= 11.00%, and N= 19.59%.

[0034] example 5 60-mol % -- in the same reaction container as the manufacture example 1 of ethoxycarbonyl-ized poly allylamine (it is  $j=0.6$ ,  $R1=H$ , and  $R2=C$  two H5 with N-allyl compound urethane system polymer I of this invention) 10.1% poly allylamine water solution (the Nitto Boseki Co., Ltd. make --) of molecular weight about 10,000 concentration PAA-10C200g (it is 0.35 mols at the monomeric unit of the poly allylamine) was put in, temperature was kept at 50 degrees C, 25.10g (a best, Nakarai Tesuku make) (0.21 mols) of diethyl carbonate was hung on the water



solution for 40 minutes, and it was dropped at it. While after dropping termination kept temperature at 50 degrees C, the reaction was continued for 24 hours. The reaction solution of transparent light yellow was obtained. pH of this solution was set to 9.55. 1g of next, this reaction solution -- a weighing bottle -- taking -- the same conditions as an example 1 -- drying -- 60-mol % -- ethoxycarbonyl-ized poly allylamine was obtained. The results of elemental analysis were C= 56.98%, H= 9.41%, and N= 13.52%. These values were in agreement with C= 57.46% of calculated value, H = 9 or 44%, and N= 13.96%.

[0035] drawing 2 -- 60-mol % -- the infrared absorption spectrum of ethoxycarbonyl-ized poly allylamine was shown. An absorption spectrum has absorption 1580cm<sup>-1</sup> based on the first amino group (- NH<sub>2</sub>), and 1700cm<sup>-1</sup> based on C=O of a urethane group (N-CO-O-R).

[0036] example 6 100-mol % -- in the same reaction container as the manufacture example 1 of ethoxycarbonyl-ized poly allylamine (it is j= 1, R1 =H, and R2 =C two H5 with N-allyl compound urethane system polymer I of this invention) 10.1% poly allylamine water solution (the Nitto Boseki Co., Ltd. make --) of molecular weight about 10,000 concentration PAA-10C200g (it is 0.35 mols at the monomeric unit of the poly allylamine) was put in, temperature was kept at 50 degrees C, 43.93g (a best, Nakarai Tesuku make) (0.37 mols) of diethyl carbonate was hung on the water solution for 1 hour, and it was dropped at it. While after dropping termination kept temperature at 50 degrees C, the reaction was continued for 24 hours. The transparent reaction solution divided into two-layer was obtained. pH of the upper layer of this solution was set to 7.81. 1g of next, lower layer reaction solutions -- a weighing bottle -- taking -- the same conditions as an example 1 -- drying -- 100-mol % -- ethoxycarbonyl-ized poly allylamine was obtained. The results of elemental analysis were C= 55.37%, H= 8.42%, and N= 10.51%. These values were in agreement with C= 55.80% of calculated value, H= 8.58%, and N= 10.84%.

[0037]

[Effect of the Invention] N-allyl compound urethane system polymer of this invention introduces the block isocyanate radical which has reactivity while changing the cation consistency of the poly allylamine system polymer by urethane-izing the amino group of the specific poly allylamine system polymer. N-allyl compound urethane system polymer of this invention is very easy to manufacture. In addition, a cation consistency and whenever [ permutation / of the block isocyanate radical which is a reaction radical ] are easily controllable. Therefore, N-allyl compound urethane system polymer of this invention offers an ingredient very effective in the field which it is the field for which current and the poly allylamine are used, and is expected a low cation consistency. Furthermore, N-allyl compound urethane system polymer of this invention offers an ingredient very effective in the field using the reactivity of a block isocyanate radical. For example, using it in the field expected the adhesive or waterproof improvement in the color binder for reactive dye, the color binder for direct dye, the anchor coat agent for an extrusion lamination, etc. is proposed.

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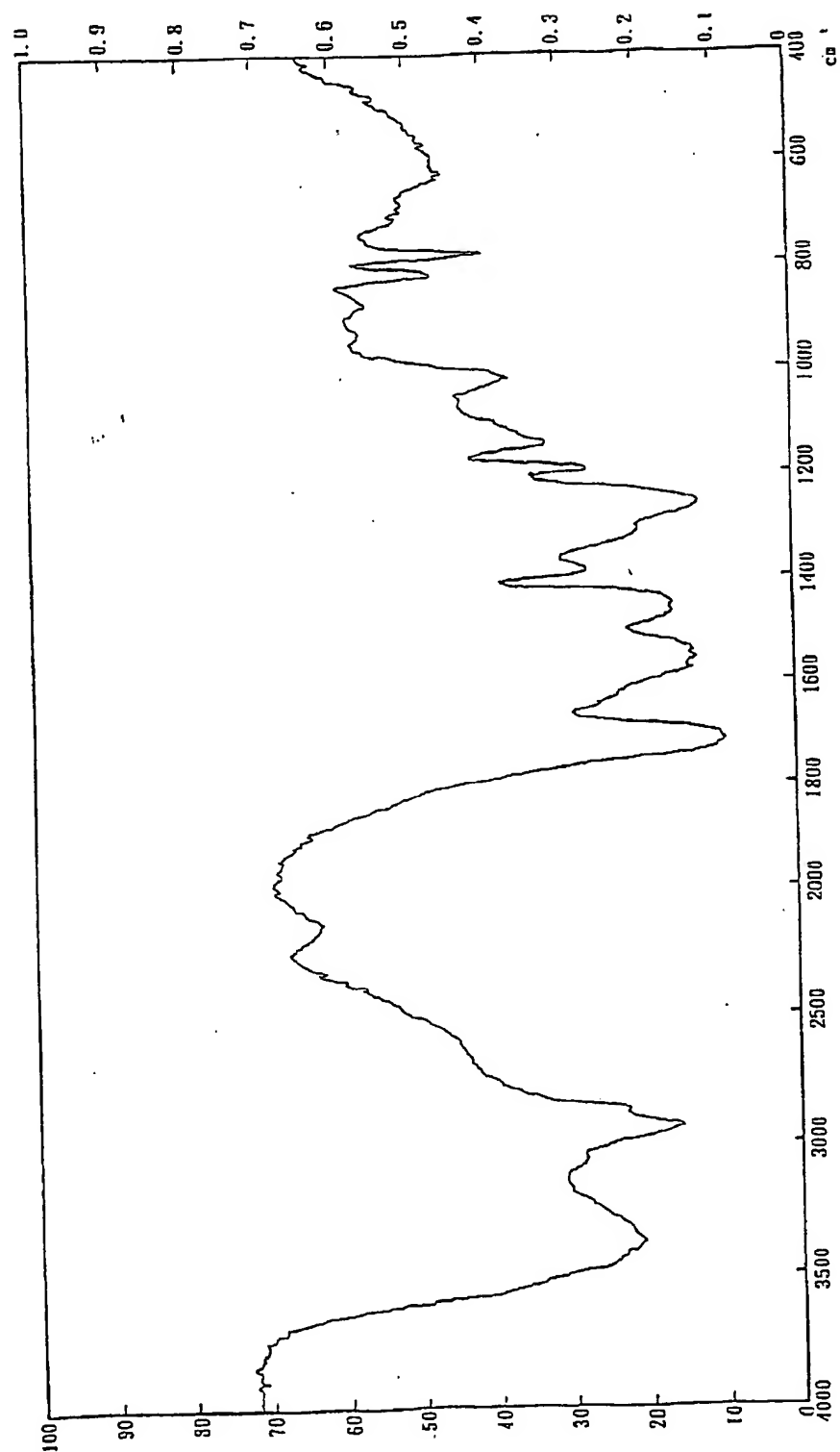
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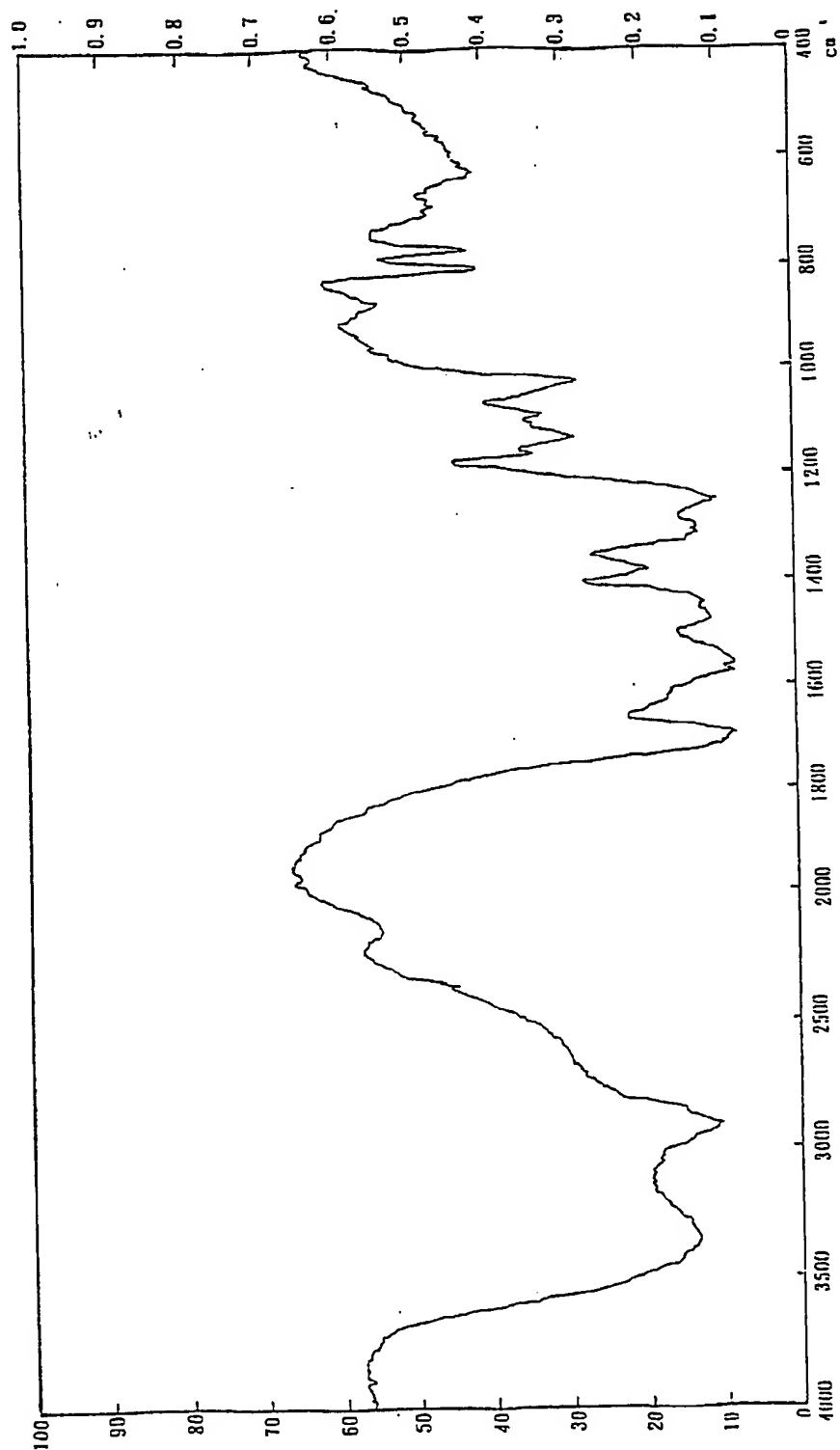
DRAWINGS

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[Drawing 1]



[Drawing 2]



[Translation done.]

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CORRECTION OR AMENDMENT

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 [Publication date] October 2, Heisei 13 (2001. 10.2)

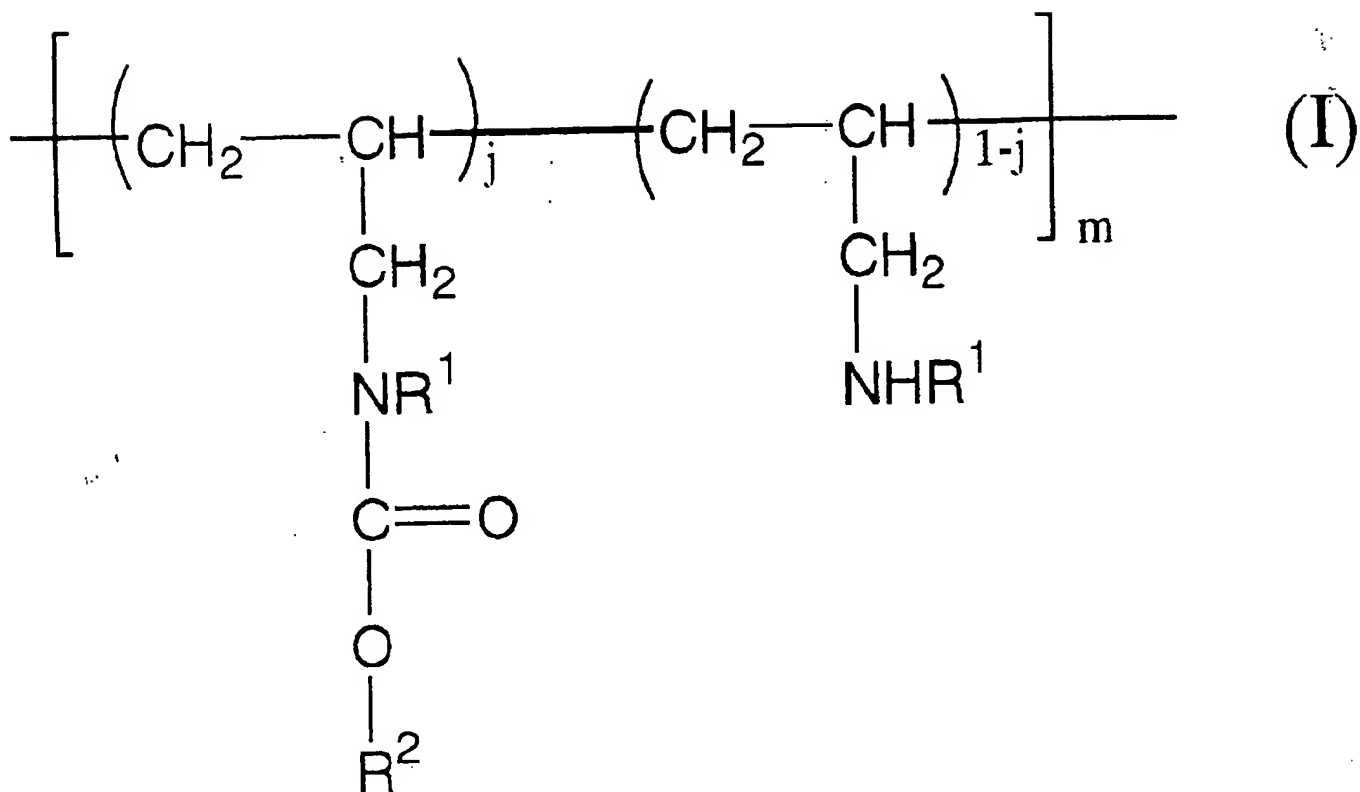
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 [Proposed Amendment]  
 [Document Name] Specification  
 [Title of the Invention] N-allyl compound urethane system polymer and its manufacture approach  
 [Claim(s)]  
 [Claim 1] General formula  
 [Formula 1]

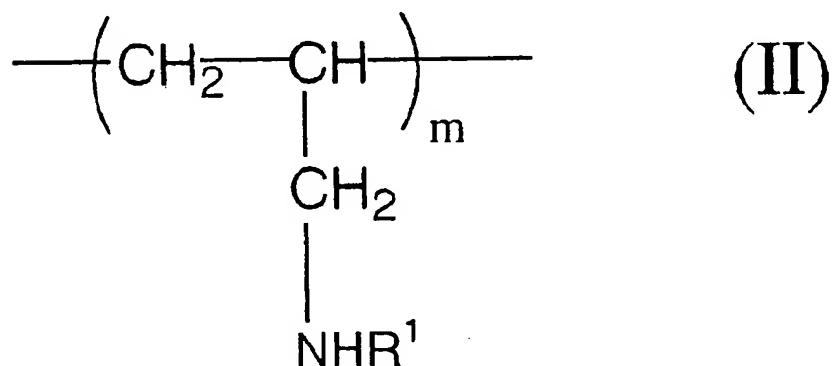


They are N-allyl compound urethane system polymer expressed with (m shows ten or more integers among a formula, j shows the number with which are satisfied of  $0 < j \leq 1$ , R<sup>1</sup> shows n- of hydrogen and carbon numbers 1-8 and an iso-alkyl group, and the substituent chosen as a list from the cycloalkyl radical of carbon numbers 5-12, and R<sup>2</sup> shows the substituent chosen from the alkyl group of carbon numbers 1-12, and the aryl group of carbon numbers 6-12), or its salt.

[Claim 2] N-allyl compound urethane system polymer according to claim 1 which R<sup>1</sup> is hydrogen and is the substituent as which R<sup>2</sup> is chosen from n- of carbon numbers 1-12, and an iso-alkyl group, or its salt.

[Claim 3] General formula

[Formula 2]



(m shows ten or more integers among a formula, and R<sup>1</sup> shows the substituent chosen as n- of hydrogen and carbon numbers 1-8, and an iso-alkyl group list from the cycloalkyl radical of carbon numbers 5-12)

In the poly allylamine system polymer come out of and expressed, or its partial salt

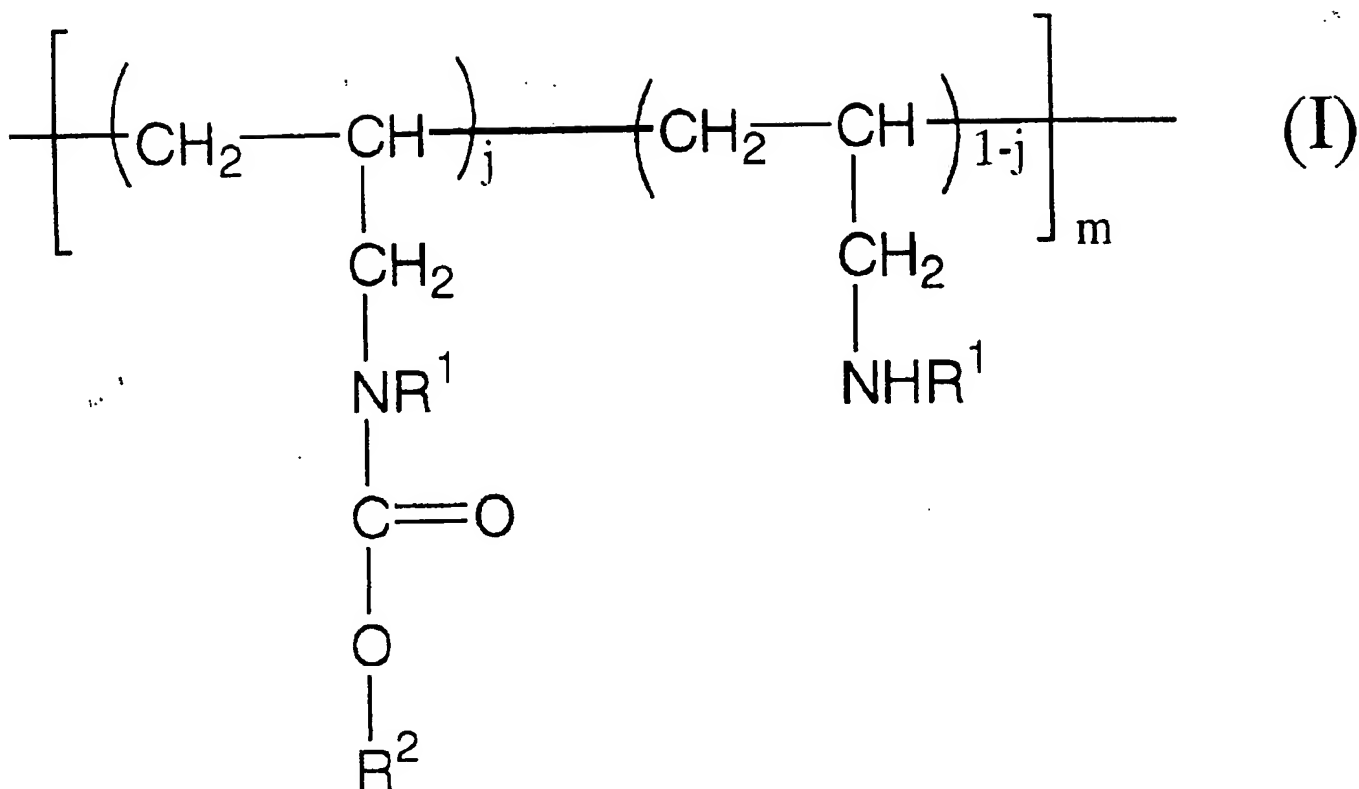
General formula

R<sup>2</sup>O-CO-OR<sup>2</sup>

(The substituent as which R<sup>2</sup> is chosen from the alkyl group of carbon numbers 1-12 and the aryl group of carbon numbers 6-12 among a formula is shown)

The general formula characterized by coming out and making the carbonic acid diester expressed react

[Formula 3]



(m shows ten or more integers among a formula, and j shows the number with which are satisfied of  $0 < j \leq 1$ .) R<sup>1</sup> shows the substituent chosen as a hydrogen list from the cycloalkyl radical of carbon numbers 5-12 by n- of carbon numbers 1-8, and the iso-alkyl group list. the substituent as which R<sup>2</sup> is chosen from the alkyl group of carbon numbers 1-12, and the aryl group of carbon numbers 6-12 -- being shown -- the manufacture approach of N-allyl compound urethane system polymer expressed or its salt.

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to new N-allyl compound urethane system polymer and its manufacture approach. Furthermore, it is related with new N-allyl compound urethane system polymer with which the amino group has a reactant side chain few in detail, and its easy manufacture approach.

[0002]

[Description of the Prior Art] The poly allylamine and a poly allylamine system polymer like Pori (N-alkyl allylamine) are cation system macromolecules which are the olefin system polymers of the straight chain containing the amino group, melt into a side chain in water well, and carry out an electric charge to plus underwater. Such a poly allylamine system polymer has peculiar reactive polymer structure and a peculiar property, therefore is used for the color binder for reactive dye, the color binder for direct dye, the food preserver, the anchor coat agent, etc. Moreover, using the poly allylamine system polymer in fields, such as silver halide photosensitive material, a sustained-release physic constituent, ion exchange resin, and functional film, is proposed. However, if it generally says as indicated by the "composition [ of a reactant polymer ], and application" 80-92 page (1989) of the CMC issue, the number of the reaction radicals of a reactive polymer does not have not much many a certain need, and even when inconvenient [, and ], there is. [ too ] Although, compounding the polymer which decreased the amino group by copolymerization on the other hand is also considered, since monoallyl amine system monomers, such as a monoallyl amine and monochrome (N-alkyl allylamine), are not copolymerized, as for the usual vinyl system monomer, it is difficult monomers to compound the poly allylamine system derivative of a low cation consistency by copolymerization. Then, how to make the amino group of the poly allylamine system polymer react with other compounds, and change into a deactivating group is also examined. The allylurea polymer (JP,63-43403,B) is known as a poly allylamine system derivative manufactured by such approach. However, the present condition is this poly allylamine system derivative's

also having problems, like it being difficult to refine, and hardly used practical.

[0003] On the other hand, since the poly allylamine system polymer melts in water well, the application of the adhesion fields, such as a drainage system anchor coat agent (JP,4-292640,A) as an alternative of an organic solvent system anchor coat agent, is known. However, there was a practical problem that a water resisting property was missing in the laminate film produced using these anchor coat agents. Therefore, development of the new poly allylamine system derivative whose water resisting property crosslinking reaction occurs easily, and insolubilizes by heat-treatment, consequently improves was desired.

[0004]

[Problem(s) to be Solved by the Invention] It is offering the poly allylamine system derivative which as for the purpose of this invention it has a reactant side chain, and crosslinking reaction's occurs easily by heat-treatment further, and can insolubilize there being little amino group, and the method of more specifically manufacturing simply N-allyl compound urethane system polymer and it.

[0005]

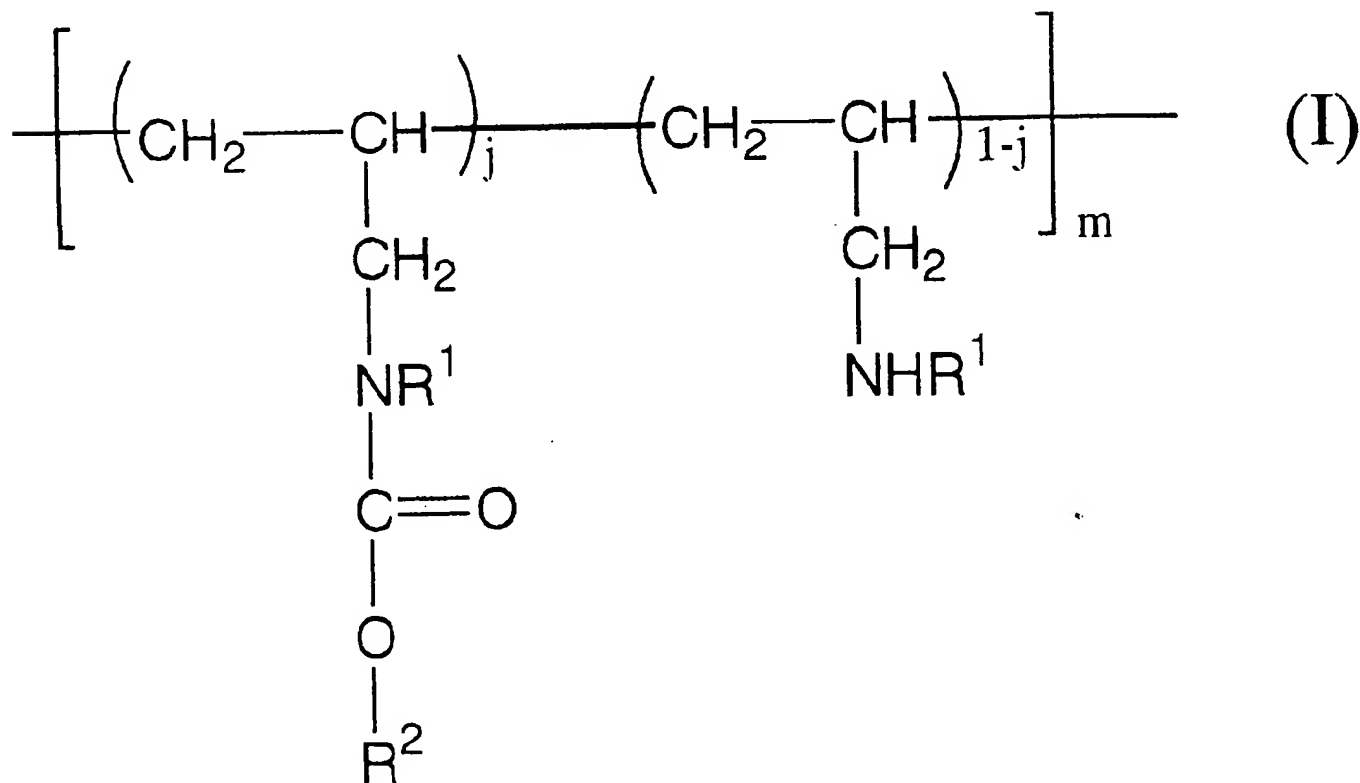
[Means for Solving the Problem] By making the specific poly allylamine system polymer and specific carbonic acid diester react, there is little amino group, and it finds out manufacturing simply new N-allyl compound urethane system polymer which has a block isocyanate radical, and this invention persons came to make this invention based on this knowledge, as a result of the amino group's repeating research wholeheartedly about the approach of manufacturing simply new N-allyl compound urethane system polymer and new it which have a reactant side chain few. In addition, on these specifications, urethane-ization means that the amino group (NH) changes to an alkyloxy carbonylamino radical or an aryloxy carbonylamino radical (N-CO-OR<sub>2</sub>).

[0006] This invention,

General formula

[0007]

[Formula 4]



They are N-allyl compound urethane system polymer expressed with (m shows ten or more integers among a formula, j shows the number with which are satisfied of  $0 < j \leq 1$ , R<sup>1</sup> shows n- of hydrogen and carbon numbers 1-8 and an iso-alkyl group, and the substituent chosen as a list from the cycloalkyl radical of carbon numbers 5-12, and R<sup>2</sup> shows the substituent chosen from the alkyl group of carbon numbers 1-12, and the aryl group of carbon numbers 6-12), or its



salt.

[0008] N-allyl compound urethane system polymer of this invention has a block isocyanate radical. Therefore, under heating conditions, since urethane becomes isocyanate, reactivity usually becomes high.

[0009] N-allyl compound urethane system polymer of this invention and its salt are the stability when making it the stability when making it a solution, especially an acidic solution, the solubility over moderate water, and the ease of receiving of a raw material to R1. Hydrogen and the thing whose R2 is n- of carbon numbers 1-12 or an iso-alkyl group are desirable.

[0010] In this invention, m expresses the polymerization degree of N-allyl compound urethane system polymer of this invention. m is 10-5000 preferably ten or more.

[0011] In this invention, j expresses whenever [ urethane-ized ]. Although j is  $0 < j \leq 1$ ,  $0.05 \leq j \leq 1$  is usually desirable from whenever [ reduction / of the cation consistency of N-allyl compound urethane system polymer of this invention ].

[0012] In this invention, R1 shows the substituent chosen as n- of hydrogen and carbon numbers 1-8, and an iso-alkyl group list from the cycloalkyl radical of carbon numbers 5-12. It is R1 practically from the ease of carrying out of the solubility over moderate water, and acquisition of a raw material. Hydrogen is desirable.

[0013] In this invention, R2 shows the substituent chosen from the alkyl group of carbon numbers 1-12, and the aryl group of carbon numbers 6-12. In this case, methyl, ethyl, and propyl can be illustrated as an alkyl group of carbon numbers 1-12. R2 can illustrate a phenyl group, o-tolyl group, m-tolyl group, p-tolyl group, etc. as an aryl group of carbon numbers 6-12. As for R2, it is desirable from the stability when making it into the stability when making it a solution, especially an acidic solution in N-allyl compound urethane system polymer of this invention that it is n-alkyl group or the iso-alkyl group of carbon numbers 1-12.

[0014] The salt of N-allyl compound urethane system polymer of this invention is an addition salt to the side-chain amino group in the monomeric unit which is not urethane-ized. As such an addition salt, hydroxy acid salts, such as carboxylate, such as a hydrochloride, the hydrobromate, an iodine hydro acid salt, a sulfate, phosphate, phosphonate, and acetate, a methansulfonic acid salt, a p-toluenesulfonic-acid salt, citrate, and a tartrate, and a benzoate can be illustrated.

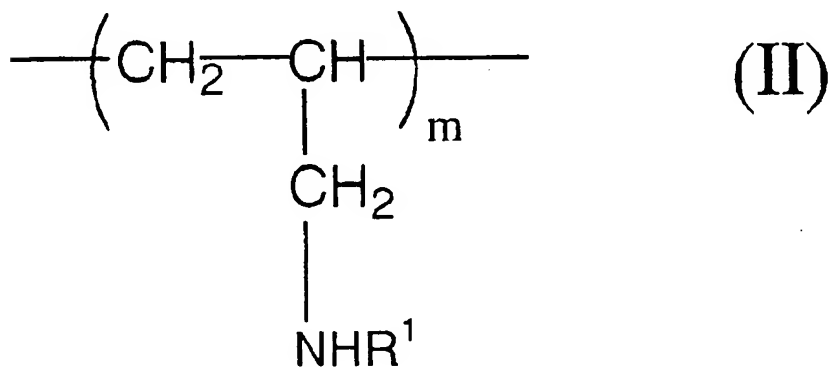
[0015] Generally, when N-allyl compound urethane system polymer of this invention is made into a salt, it may become unstable in the state of a solution. Therefore, in the state of a solution, the free thing of N-allyl compound urethane system polymer of this invention is desirable.

[0016]

[Embodiment of the Invention] N-allyl compound urethane system polymer of this invention can make the carbonic acid diester of the specific poly allylamine system polymer and specification able to react, and can be manufactured. That is, N-allyl compound urethane system polymer of this invention is a general formula.

[0017]

[Formula 5]



It is the poly allylamine system polymer expressed with (m shows ten or more integers among a formula, and R1 shows n- of hydrogen and carbon numbers 1-8 and an iso-alkyl group, and the substituent chosen as a list from the cycloalkyl radical of carbon numbers 5-12),

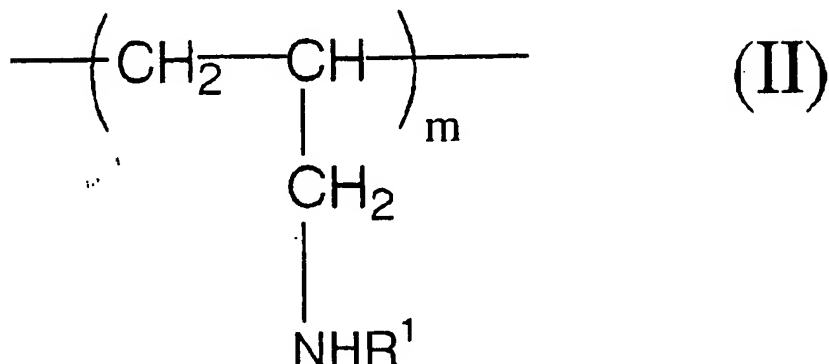
General formula

R2O-CO-OR2

It can manufacture by making the carbonic acid diester expressed with (the substituent as which R2 is chosen from the alkyl group of carbon numbers 1-12 and the aryl group of carbon numbers 6-12 among a formula is shown) react. The poly allylamine system polymer of a raw material for manufacturing N-allyl compound urethane system polymer of this invention is a general formula.

[0018]

[Formula 6]

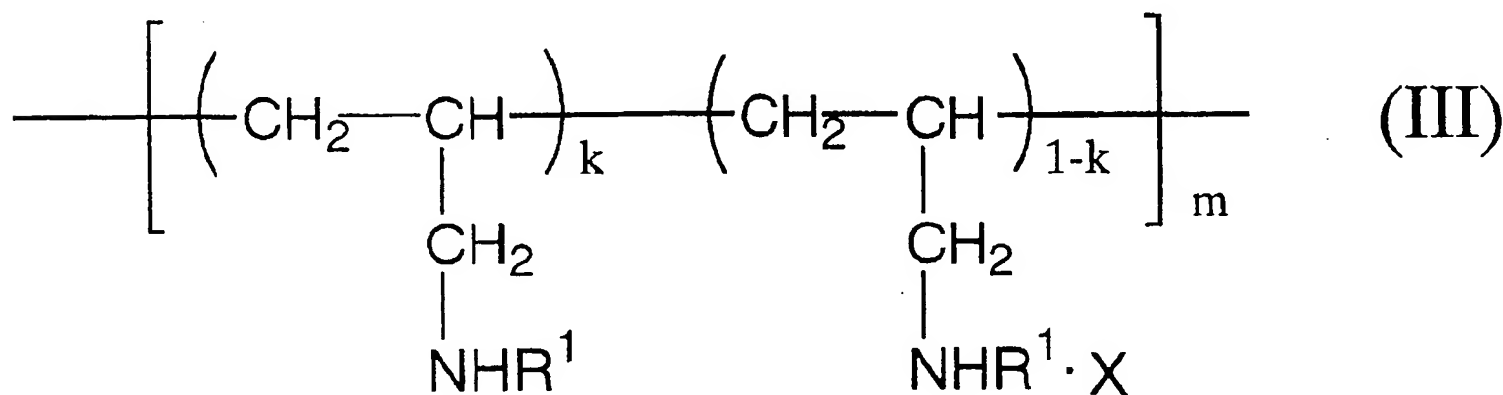


It is expressed with (m shows ten or more integers among a formula, and R1 shows n- of hydrogen and carbon numbers 1-8 and an iso-alkyl group, and the substituent chosen as a list from the cycloalkyl radical of carbon numbers 5-12).

[0019] Moreover, the partial salt of the poly allylamine system polymer can also be used as such a raw material. Such a partial salt,

[0020]

[Formula 7]



It is expressed with (being a salt m showing ten or more integers among a formula, for k showing the number with which are satisfied of  $0 < k < 1$ , for R1 showing n- of hydrogen and carbon numbers 1-8 and an iso-alkyl group, and the substituent chosen as a list from the cycloalkyl radical of carbon numbers 5-12, and for X forming a partial salt). If it is the acid which can form the amino group and salt of a side chain of the poly allylamine system polymer as an acid for forming this partial salt, it will not limit especially. As such an acid, hydroxy acid, such as carboxylic acids, such as a hydrochloric acid, a hydrobromic acid, iodine hydro acid, a sulfuric acid, a phosphoric acid, phosphonic acid, and an acetic acid, methansulfonic acid, p-toluenesulfonic acid, a citric acid, and a tartaric acid, and a benzoic acid can be illustrated.

[0021] The poly allylamine system polymer of a raw material is the poly allylamine when R1 is hydrogen. The free type thing of the poly allylamine is desirable. Although what neutralized the salt of the known poly allylamine with alkali can be used as a free type the poly allylamine (molecular weight 10,000 [ about ]) water solution (the Nitto Boseki Co., Ltd. make --) of 15% of commercial concentration PAA-15 and the poly allylamine (molecular weight 10,000 [ about ]) water solution (the Nitto Boseki Co., Ltd. make --) of 10% of concentration PAA-10C, the poly

allylamine (molecular weight 10,000 [ about ]) water solution (the Nitto Boseki Co., Ltd. make, PAA-L) of 20% of concentration, the poly allylamine (molecular weight 100,000 [ about ]) water solution (the Nitto Boseki Co., Ltd. make, PAA-H) of 20% of concentration, etc. may be used as it is.

[0022] When R1 is n- of carbon numbers 1-8 and an iso-alkyl group, and the substituent chosen as a list from the cycloalkyl radical of carbon numbers 5-12 in the poly allylamine system polymer of a raw material By approach which is indicated by JP,63-43402,B, JP,6-2779,B, and JP,2-57084,B The salt of Pori (N-alkyl allylamine) can be manufactured and what removed the neutralization salt which subsequently carries out the byproduction of the water solution of the salt of the Pori (N-alkyl allylamine) after neutralizing with alkali, for example, a sodium hydroxide, for example, a sodium chloride, by dialysis can be used.

[0023] As a solvent for using the poly allylamine system polymer of a raw material as a solution, the mixed solvent of water, an organic solvent, or a water and an organic solvent can be used. As an organic solvent, the solubility of a raw material to a polar solvent is desirable, and alcohols, such as a methanol, ethanol, 1-propanol, 2-propanol, and 1-butanol, an acetonitrile, a formamide, N,N-dimethylformamide, dimethyl sulfoxide, a tetrahydrofuran, 1,4-dioxane, etc. can be illustrated. As for the amount of the solvent used for using the poly allylamine system polymer of a raw material as a solution, it is desirable that it is 0.5 to 100 weight to poly allylamine system polymer 1 weight of a raw material.

[0024] The carbonic acid diester of a raw material for manufacturing N-allyl compound urethane system polymer of this invention is expressed with general formula R2 O-CO-OR2 (R2 shows among a formula the substituent chosen from the alkyl group of carbon numbers 1-12, and the aryl group of carbon numbers 6-12). As carbonic acid diester, dimethyl carbonate, diethyl carbonate, diphenyl carbonate, etc. can be illustrated. The amount of the carbonic acid diester made to react can usually use required computational complexity for whenever [ urethane-ized / of N-allyl compound urethane system polymer ] at stoichiometric.

[0025] When making the poly allylamine system polymer and carbonic acid diester of a raw material react and manufacturing N-allyl compound urethane system polymer of this invention, it is good to drop carbonic acid diester at the solution of the poly allylamine system polymer of a raw material slowly first. At this time, carbonic acid diester can be dissolved in a solvent and it can also be dropped at the solution of the poly allylamine system polymer of a raw material. In this case, the solvent for dissolving carbonic acid diester is usually the same as the solvent for dissolving the poly allylamine system polymer of a raw material. As for the reaction of the poly allylamine system polymer and carbonic acid diester, it is desirable to carry out agitating. Reaction temperature is preferably good to maintain [ 0-100-degree C ] at 30-60 degrees C still more preferably. When reaction temperature is too high, the generated urethane may decompose. Reaction time is 12 - 25 hours preferably, and can usually obtain the solution of the allyl compound urethane system polymer of this invention for 12 to 48 hours. In order to remove the alcohol and the reaction solvent which carried out the byproduction after reaction termination, N-allyl compound urethane system polymer of this invention can be obtained as a solid-state by carrying out the vacuum drying of the reaction solution. When dimethyl carbonate, diethyl carbonate, or carbonic acid dipropyl is used as water and carbonic acid diester as a solvent, as for a vacuum drying, it is desirable that temperature carries out by 25-70 degrees C and the vacua desirable and suitable at 35-60 degrees C. When temperature is too high, side reaction may start. Moreover, the salt of N-allyl compound urethane system polymer of this invention can manufacture this and carbonic acid diester by making it react like the case where the free poly allylamine system polymer is used, using the partial salt of the poly allylamine system polymer as a raw material. usually, the case where the partial salt and carbonic acid diester of the poly allylamine system polymer of a raw material are made to react -- the poly allylamine system polymerization -- NH which does not form the salt is preferentially urethane-ized by NH in the living body. It becomes possible to take out the salt of N-allyl compound urethane system polymer of this invention as a solid-state by adding the solution of the salt of N-allyl compound urethane system polymer obtained to solvents, such as an acetone, and reprecipitating it after reaction termination.

[0026] It depends for whenever [ urethane-ized / of N-allyl compound urethane system polymer of this invention ] (mol %) on the amount of the carbonic acid diester of the used raw material. When the carbonic acid diester of an equimolecular amount is used to the amino group of the poly allylamine system polymer of a raw material, the amino group is usually almost urethane-ized. Therefore, the cation consistency of N-allyl compound urethane system polymer of this invention can be adjusted by adjusting the amount of the carbonic acid diester used as a raw material. Moreover, the hydrophobicity of N-allyl compound urethane system polymer of this invention can be changed according to the class of carbonic acid diester to be used. Therefore, when using N-allyl compound urethane system polymer of this

invention for various applications, what has a suitable cation consistency and has suitable hydrophobicity can be chosen.

[0027] Furthermore, N-allyl compound urethane system polymer of this invention can choose the dissociation temperature of a block isocyanate radical by choosing the urethane group to introduce from being also the poly allylamine which has a block isocyanate radical.

[0028] Below, an example shows N-allyl compound urethane system polymer and its manufacture approach of this invention. As for this invention, it is needless to say that it is not what is limited by these examples.

[0029] example 1 20-mol % -- manufacture of methoxycarbonyl-ized poly allylamine (they are  $j=0.2$ ,  $R_1=H$ , and  $R_2=CH_3$  with N-allyl compound urethane system polymer I of this invention)

200g (the Nitto Boseki Co., Ltd. make, PAA-10C) (it is 0.35 mols at the monomeric unit of the poly allylamine) of the poly allylamine (molecular weight 10,000 [ about ]) water solutions of 10.1% of concentration was put into the 300ml separable flask equipped with the agitator, the tap funnel, the thermometer, and the reflux condenser, temperature was kept at 50 degrees C, 6.38g (product made from MITEKKUSU) (0.07 mols) of dimethyl carbonate was hung on the water solution for 15 minutes, and it was dropped at it. While after dropping termination kept temperature at 50 degrees C, the reaction was continued for 12 hours and the reaction solution of transparent light yellow was obtained. pH of this solution was 10.84. next, the desiccating agent after taking 1g of this reaction solution to a weighing bottle and carrying out a vacuum drying at a room temperature for 24 hours -- phosphoric oxide -- using -- 50 degrees C -- a 48-hour vacuum drying -- carrying out -- 20-mol % -- methoxycarbonyl-ized poly allylamine was obtained. The results of elemental analysis were C= 58.97%, H= 10.93%, and N= 19.76%. These values were mostly in agreement with C= 59.44% of calculated value, H= 10.86%, and N= 20.39%.

[0030] example 2 60-mol % -- manufacture of methoxycarbonyl-ized poly allylamine (they are  $j=0.6$ ,  $R_1=H$ , and  $R_2=CH_3$  with N-allyl compound urethane system polymer I of this invention)

Into the same reaction container as an example 1, 200g (the Nitto Boseki Co., Ltd. make, PAA-10C) (it is 0.35 mols at the monomeric unit of the poly allylamine) of the poly allylamine (molecular weight 10,000 [ about ]) water solutions of 10.1% of concentration was put in, temperature was kept at 50 degrees C, 19.14g (product made from MITEKKUSU) (0.21 mols) of dimethyl carbonate was hung on the water solution for 40 minutes, and it was dropped at it. While after dropping termination kept temperature at 50 degrees C, the reaction was continued for 12 hours and the reaction solution of transparent light yellow was obtained. pH of this solution was 9.78. 1g of next, this reaction solution -- a weighing bottle -- taking -- the same conditions as an example 1 -- drying -- 60-mol % -- methoxycarbonyl-ized poly allylamine was obtained. The results of elemental analysis were C= 54.32%, H= 9.17%, and N= 14.75%. These values were mostly in agreement with C= 54.88% of calculated value, H= 8.99%, and N= 15.24%.

[0031] drawing 1 -- 60-mol % -- KBr of methoxycarbonyl-ized poly allylamine -- law -- the infrared absorption spectrum was shown. Absorption of 1580cm<sup>-1</sup> based on the first amino group (-NH<sub>2</sub>) and 1700cm<sup>-1</sup> based on C=O of a urethane group (N-CO-O-R) was accepted in the absorption spectrum.

[0032] example 3 100-mol % -- manufacture of methoxycarbonyl-ized poly allylamine (they are  $j=1$ ,  $R_1=H$ , and  $R_2=CH_3$  with N-allyl compound urethane system polymer I of this invention)

Into the same reaction container as an example 1, 200g (the Nitto Boseki Co., Ltd. make, PAA-10C) (it is 0.35 mols at the monomeric unit of the poly allylamine) of the poly allylamine (molecular weight 10,000 [ about ]) water solutions of 10.1% of concentration was put in, temperature was kept at 50 degrees C, 33.50g (product made from MITEKKUSU) (0.37 mols) of dimethyl carbonate was hung on the water solution for 1 hour, and it was dropped at it. While after dropping termination kept temperature at 50 degrees C, the reaction was continued for 12 hours and the reaction solution of transparent light yellow was obtained. pH of this solution was 7.85. 1g of next, this reaction solution -- a weighing bottle -- taking -- the same conditions as an example 1 -- drying -- 100-mol % -- methoxycarbonyl-ized poly allylamine was obtained. The results of elemental analysis were C= 51.68%, H= 7.94%, and N= 11.83%. These values were mostly in agreement with C= 52.16% of calculated value, H= 7.88%, and N= 12.17%.

[0033] example 4 20-mol % -- manufacture of ethoxycarbonyl-ized poly allylamine (it is  $j=0.2$ ,  $R_1=H$ , and  $R_2=C_2H_5$  with N-allyl compound urethane system polymer I of this invention)

Into the same reaction container as an example 1, 200g (the Nitto Boseki Co., Ltd. make, PAA-10C) (it is 0.35 mols at the monomeric unit of the poly allylamine) of the poly allylamine (molecular weight 10,000 [ about ]) water solutions

of 10.1% of concentration was put in, temperature was kept at 50 degrees C, 8.37g (a best, Nakarai Tesuku make) (0.07 mols) of diethyl carbonate was hung on the water solution for 15 minutes, and it was dropped at it. While after dropping termination kept temperature at 50 degrees C, the reaction was continued for 24 hours and the reaction solution of transparent light yellow was obtained. pH of this solution was 10.88. 1g of next, this reaction solution -- a weighing bottle -- taking -- the same conditions as an example 1 -- drying -- 20-mol % -- ethoxycarbonyl-ized poly allylamine was obtained. The results of elemental analysis were C= 59.94%, H= 10.79%, and N= 19.32%. These values were mostly in agreement with C= 60.47% of calculated value, H= 11.00%, and N= 19.59%.

[0034] example 5 60-mol % -- manufacture of ethoxycarbonyl-ized poly allylamine (it is  $j = 0.6$ ,  $R_1 = H$ , and  $R_2 = C_2H_5$  with N-allyl compound urethane system polymer I of this invention)

Into the same reaction container as an example 1, 200g (the Nitto Boseki Co., Ltd. make, PAA-10C) (it is 0.35 mols at the monomeric unit of the poly allylamine) of the poly allylamine (molecular weight 10,000 [ about ]) water solutions of 10.1% of concentration was put in, temperature was kept at 50 degrees C, 25.10g (a best, Nakarai Tesuku make) (0.21 mols) of diethyl carbonate was hung on the water solution for 40 minutes, and it was dropped at it. While after dropping termination kept temperature at 50 degrees C, the reaction was continued for 24 hours and the reaction solution of transparent light yellow was obtained. pH of this solution was 9.55. 1g of next, this reaction solution -- a weighing bottle -- taking -- the same conditions as an example 1 -- drying -- 60-mol % -- ethoxycarbonyl-ized poly allylamine was obtained. The results of elemental analysis were C= 56.98%, H= 9.41%, and N= 13.52%. These values were mostly in agreement with C= 57.46% of calculated value, H = 9 or 44%, and N= 13.96%.

[0035] drawing 2 -- 60-mol % -- the infrared absorption spectrum of ethoxycarbonyl-ized poly allylamine was shown. Absorption was accepted in the absorption spectrum by 1580cm<sup>-1</sup> based on the first amino group (- NH<sub>2</sub>), and 1700cm<sup>-1</sup> based on C=O of a urethane group (N-CO-O-R).

[0036] example 6 100-mol % -- manufacture of ethoxycarbonyl-ized poly allylamine (it is  $j = 1$ ,  $R_1 = H$ , and  $R_2 = C_2H_5$  with N-allyl compound urethane system polymer I of this invention)

Into the same reaction container as an example 1, 200g (the Nitto Boseki Co., Ltd. make, PAA-10C) (it is 0.35 mols at the monomeric unit of the poly allylamine) of the poly allylamine (molecular weight 10,000 [ about ]) water solutions of 10.1% of concentration was put in, temperature was kept at 50 degrees C, 43.93g (a best, Nakarai Tesuku make) (0.37 mols) of diethyl carbonate was hung on the water solution for 1 hour, and it was dropped at it. While after dropping termination kept temperature at 50 degrees C, the reaction was continued for 24 hours and the transparent reaction solution divided into two-layer was obtained. pH of the upper layer of this solution was 7.81. 1g of next, lower layer reaction solutions -- a weighing bottle -- taking -- the same conditions as an example 1 -- drying -- 100-mol % -- ethoxycarbonyl-ized poly allylamine was obtained. The results of elemental analysis were C= 55.37%, H= 8.42%, and N= 10.51%. These values were mostly in agreement with C= 55.80% of calculated value, H= 8.58%, and N= 10.84%.

[0037]

[Effect of the Invention] N-allyl compound urethane system polymer of this invention changes the cation consistency of the poly allylamine system polymer by urethane-izing the amino group of the specific poly allylamine system polymer. Therefore, N-allyl compound urethane system polymer of this invention offers an ingredient very effective in the field which it is the field for which current and the poly allylamine are used, and is expected a low cation consistency. Moreover, since N-allyl compound urethane system polymer of this invention introduces the block isocyanate radical which has reactivity, it offers an ingredient very effective in the field using the reactivity of a block isocyanate radical. For example, using it in the field expected the adhesive or waterproof improvement in the color binder for reactive dye, the color binder for direct dye, the anchor coat agent for an extrusion lamination, etc. is proposed. N-allyl compound urethane system polymer of this invention is very easy to manufacture. In addition, a cation consistency and whenever [ permutation / of the block isocyanate radical which is a reaction radical ] are easily controllable.

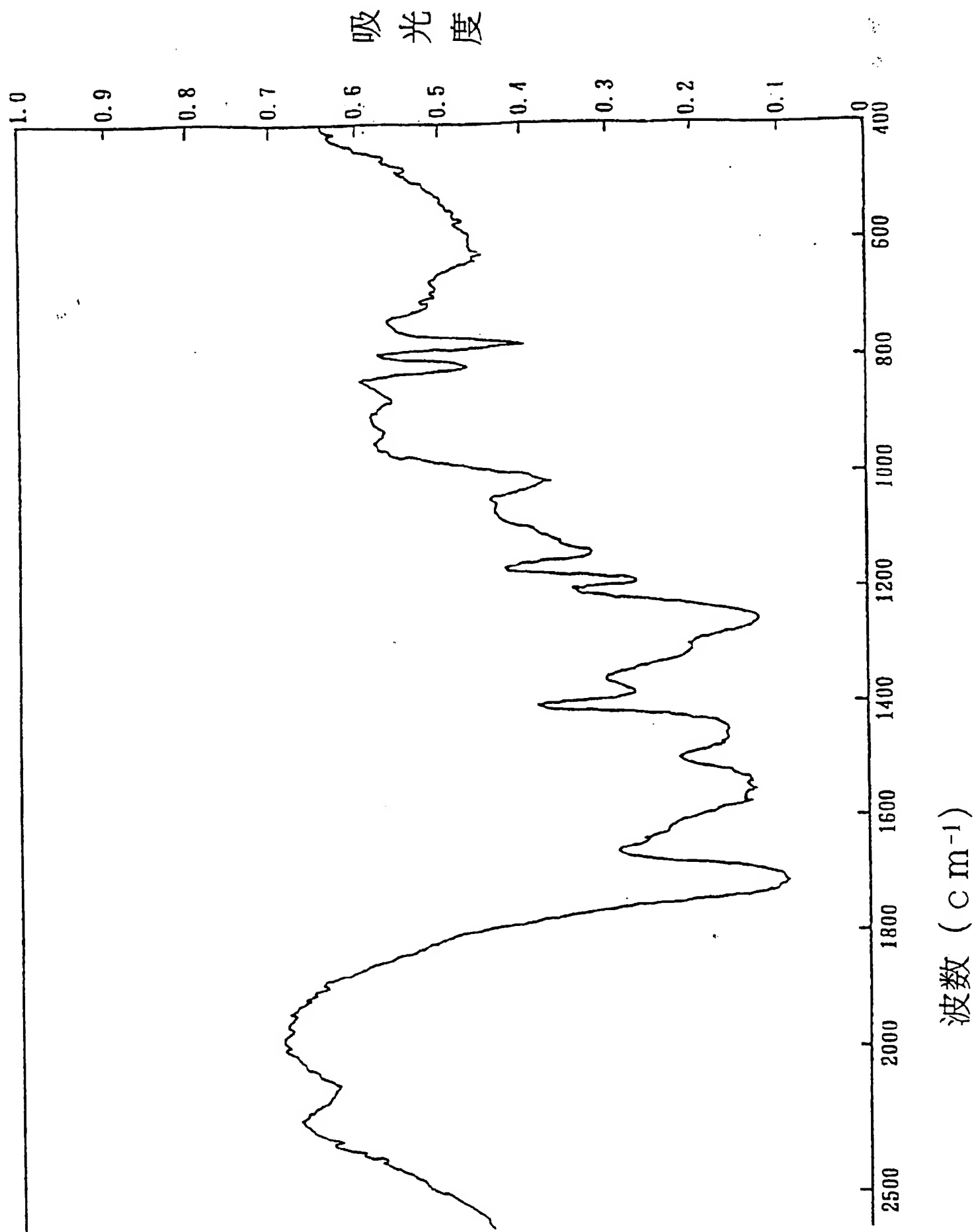
[Brief Description of the Drawings]

[Drawing 1] 60-mol % -- the infrared absorption spectrum of methoxycarbonyl-ized poly allylamine (N-allyl compound urethane system polymer of an example 2) is shown.

[Drawing 2] 60-mol % -- the infrared absorption spectrum of ethoxycarbonyl-ized poly allylamine (N-allyl compound urethane system polymer of an example 5) is shown. An axis of abscissa expresses the wave number (cm<sup>-1</sup>), and an axis of ordinate expresses permeability (%) or an absorbance.

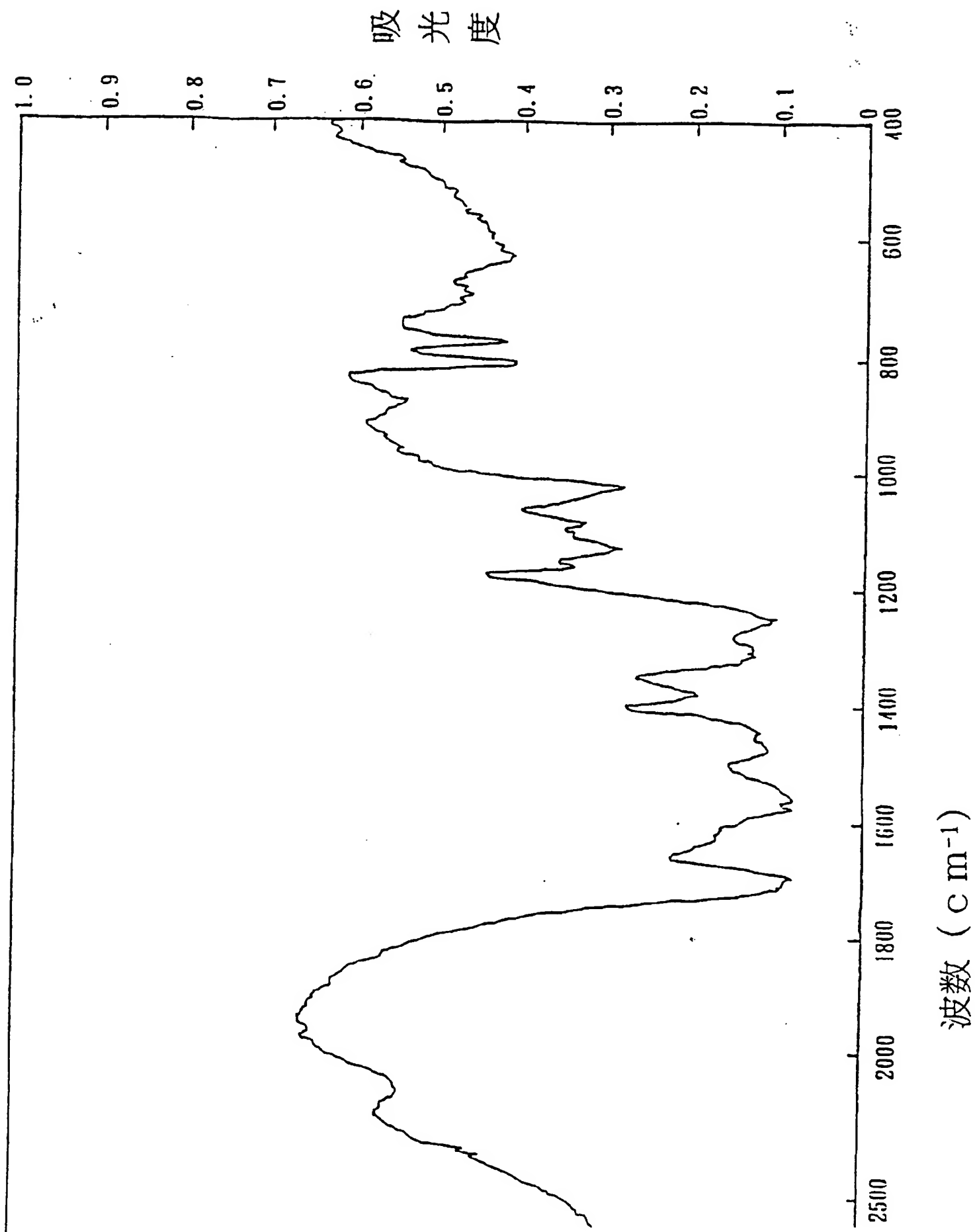
[Procedure amendment 2]

[Document to be Amended] DRAWINGS  
[Item(s) to be Amended] Complete diagram  
[Method of Amendment] Modification  
[Proposed Amendment]  
[Drawing 1]



[Drawing 2]





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[Translation done.]